

Dielectric and Piezoelectric Performance of Soft PZT Piezoceramics Under Simultaneous Alternating Electromechanical Loading

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Abstract

This study is focused on the investigation of the fundamental properties of piezoceramics under loading conditions simulating the in-service environment of high-strain actuators. High electric field induced polarization and strain responses were experimentally evaluated for a commercial soft PZT material subjected to cyclic mechanical load with different mean stresses and amplitudes. When the stress was applied in-phase with electrical loading, the polarization and strain outputs are found to monotonically decrease with an increase in stress amplitude, until mechanical loading completely impedes the piezoelectric response. An inverse effect occurs for the out-of-phase electromechanical loading tests, in which the polarization and strain outputs increase with increasing stress amplitude. In general, the enhanced polarization and strain responses are accompanied by an unfavorable increased hysteresis and nonlinearity. An attempt has been made to explain the experimental findings by simultaneously taking into account the effects of elastic deformation, domain reorientation, and piezoeffects.

1. Introduction

Piezoelectric actuators offer many excellent characteristics that are attractive to smart structure and system applications, such as large generative force, quick response time, low power consumption, and compactness.^{1, 2} When composed in the vicinity of the morphotropic phase boundary (MPB), lead zirconate titanate (PZT) ceramics and related modified families exhibit

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outstandingly high electromechanical coupling coefficients which make them the most widely commercialized and studied piezoelectric materials.^{3,4} Polycrystalline piezoceramics are subdivided into domains separated by domain walls. Application of an electric field or a mechanical load in excess of a critical magnitude may reorient the spontaneous polarization direction of domains, which is commonly called ferroelectric or ferroelastic domain switching. The consequence of domain switching is the occurrence of a non-linear hysteresis behavior.^{5,6} For actuators used in practice, the simplest but frequently encountered loading spectrum is uniaxial electromechanical loading, i.e. the electric field and a co-axial stress are applied to a component along its poling axis (3-direction). The resulting electric displacement (D_3) and longitudinal strain (S_3) can be described as

$$D_3 = \epsilon_{33}E_3 + P_3^r + d_{33}\sigma_3 \quad (1)$$

$$S_3 = s_{33}\sigma_3 + S_3^r + d_{33}E_3 \quad (2)$$

where $\epsilon_{33}E_3$ and $s_{33}\sigma_3$ represent the dielectric contribution and mechanical elastic strain, P_3^r and S_3^r correspond to the domain switching-related irreversible remnant polarization and strain, and $d_{33}\sigma_3$ (direct) as well as $d_{33}E_3$ (inverse) are the contributions of piezoeffects. Note that the dielectric and piezoelectric coefficients are functions of the poling state, which in turn depends on the electric field and stress loading conditions.

Piezoceramics are widely used in a multitude of ultrasonic applications, like buzzers, surface acoustic wave filters, hydrophone, and ultrasonic cleaners, etc.² Due to the facts that they are operating under high-frequency (resonance) and small-signal loading, domain switching related irreversible contributions (P_3^r and S_3^r) in Eq. 1-2 can be omitted, the dielectric and actuation (strain) responses are therefore governed by linear behaviour. The material's parameters provided by the manufacturers are normally determined by weak-field measurements performed using a resonance-antiresonance method.⁷

Considerable efforts were made in recent years to use piezoceramics for low-frequency (off-resonance) novel actuator applications, where high forces and/or large displacement output are required. Some of the most promising industrial applications include precision positioning, vibration suppression devices, noise controls, and fast fuel injection valves in automotive areas.^{8,9} Piezoelectric materials offer excellent capability of high force generation, but the actuation displacements are limited. To obtain high strain output, novel designs, e.g. multilayer stacks and mechanical amplification devices, are utilized; furthermore, the active material has to be subjected to high driving electric field.¹⁰

The mechanical preloading mechanism (or prestress) is an essential component of the actuator design. It is well known that the tensile strength of piezoceramics is much lower than the compressive strength.¹¹ Therefore, the actuators are normally designed to operate under significant compressive preload to avoid failure. Superimposition of a compressive stress can effectively prevent premature delamination and cracking from occurring in multilayer stacks.

Large signal external loads lead to ferroelectric and/or ferroelastic domain switching in the material. Due to the change of remnant polarization and strain (poling state), the material's parameters in Eq.1-2 are no longer constants, but depend on the history of applied signals. As a result, the real response of piezoceramics will display significant hysteresis and non-linearity.

Faced with the design, working condition optimization, and especially reliability assessment of novel actuators requiring high force and/strain output, some research efforts have been made to investigate experimentally the dielectric and actuation behavior of bulk ceramics or piezo-stacks under large-signal combined electro-mechanical loading conditions.^{5,9,12-14}

Enhanced actuation capabilities associated with larger nonlinearity and hysteresis were confirmed for a component driven by a high electric field within a precise compression preload range. This phenomenon is generally attributed to extra extrinsic contribution of more non-180° domain switching induced by the combined action of electro-mechanical loading.

These studies provided valuable information, however, most of the measurements were simply performed under a constant (dead) compression preload. For applications in practice, the actuators may be designed to operate under cyclic stress loading. That is, the active material will experience both electrical and mechanical loading at the same time. Knowledge of material's properties under more complicated loading spectra is desirable. A preliminary work was carried out by Mitrovic et al.⁹, in which only the strain behavior was investigated for piezoelectric stacks upon the application of simultaneous cyclic electromechanical load and, especially, the compressive stress was applied in step with electric field.

In this paper, we evaluate the response of a commercial piezoceramic under combined electro-mechanical loading as it relates to the in-service condition. High electric field induced polarization and strain measurements were carried out under alternating mechanical load of various mean stresses and amplitudes, which was applied in-phase as well as out-of-phase with electrical loading. The experimental findings were explained by simultaneously taking into account the contributions of elastic deformation, domain switching, and piezoeffects.

2. Experimental procedures

2.1. Materials, specimen preparation, and experimental setup

Measurements were performed on the commercially available PIC 151 “soft” PZT piezoceramic (PI Ceramic, Lederhose, Germany). The samples were cut and ground to rectangular blocks of $5 \times 5 \times 15 \text{ mm}^3$ by the manufacturer, with silver electrodes burned into the upper and bottom $5 \times 5 \text{ mm}^2$ surfaces. Following stringent cleaning, four strain gauges were attached to the sample using an M-bond 600 adhesive (Vishay Measurements Group, Raleigh, CA). A pair of oppositely mounted gauges was used to measure the longitudinal strain, another two were employed for transverse strain monitoring.

The prepared specimen was installed into a carefully designed test fixture capable of simultaneously applying uniaxial compressive stress and electric field. The top test fixture incorporated a spherical joint assembly to accommodate slight misalignment. An Instron servohydraulic load frame was used to apply mechanical loads. The high E field was provided by a bipolar high-voltage power supply (HCB 15–30 000, F. u. G., Rosenheim, Germany) with a maximum output of ± 30 kV. During measurement, the sample was immersed in an insulated electric liquid (FC-40, 3M, St. Paul, MN) to prevent arcing. More details about the specimen preparation and the testing setup can be found in Refs. 14,15.

Sawyer-Tower method was applied to measure the electric polarization using a high-input-resistance electrometer (6517A, Keithley Instruments, Cleveland, OH).

KWS 3073 5 kHz carrier frequency amplifiers (HBM, Darmstadt, Germany) were used to monitor the strains. Extra caution was taken to minimize the bending effects. Upon the application of a very small compression load, the strain readings from each pair of oppositely attached strain gauges were compared to detect the degree of misalignment introduced in the specimen. The sample position was then adjusted repeatedly to ensure the stress applied along the central axis. Furthermore, the data recorded by each pair of gauges were averaged to plot the curves after the experiments.

A computer equipped with a data acquisition board and running DASYPALAB software (Dasytec, Amherst, NH) was used to digitally record the measurement signals and control the Instron machine as well as the high-voltage power supply.

2.2. *Measurement procedure*

The initially unpoled specimen was first poled by full electric field reversals (± 2 kV/mm, 4 cycles) in the stress-free state. After poling, ramp-shape cyclic electric field with a range limited between -0.4 and $+2$ kV/mm was applied on the specimen to conduct the designed

measurements. The coercive field (E_c) of this material is about ± 1 kV/mm, the selected field range is the maximum recommended for high-strain actuator applications. For all the measurement, the field loading rate is fixed to $0.08 \text{ kV}\cdot\text{mm}^{-1}\cdot\text{s}^{-1}$.

Two series of combined electro-mechanical tests were designed to simulate the practical operation of piezo-actuators assuming the cyclic stress and electric field loading occurred at the same frequency. The mechanical load was rigidly restricted in compressive range. In each stress state with a certain amplitude, a total of five E field loading cycles were applied, and only the stable responses induced in the last cycle were used to plot the curves.

In the first series of tests, the maximum stress σ_{\max} is zero. That is, the stress ranges from zero down to some compressive minimum and then back to zero. In this special case the mean stress is half the minimum stress, or $\sigma_m = \sigma_{\min}/2$. Such a stress-time pattern, often referred to as *released compression* in the literature, is illustrated in Fig. 1 for a stress range ($\Delta\sigma = \sigma_{\max} - \sigma_{\min}$) of 25 MPa as an example.

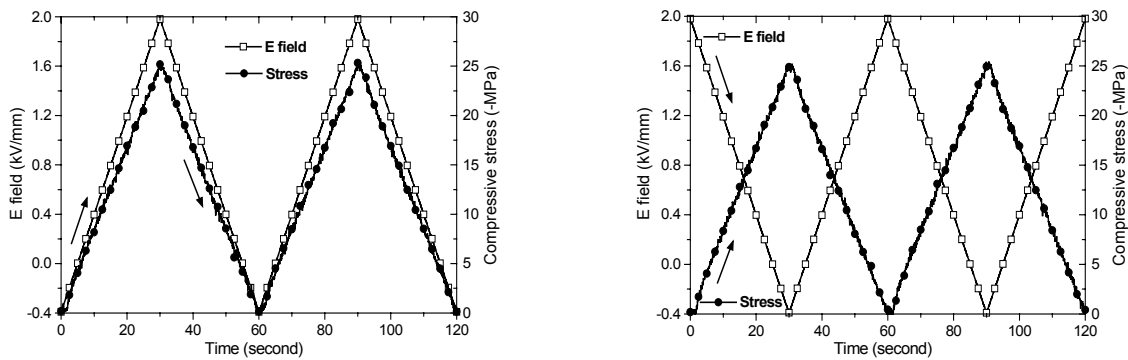


Fig. 1. Electric field-time and stress-time patterns in which the maximum stress is zero. (Left) In-phase loading. (Right) Out-of-phase loading.

In the second series of tests, the stress spectrum to which the specimen is subjected is a constant-mean stress-time pattern of various amplitudes. Such a constant-mean case may be thought of as a static stress equal in magnitude to the mean σ_m , with a superposed completely reversed cyclic stress of amplitude σ_a ($\sigma_a = (\sigma_{\max} - \sigma_{\min})/2$).

To simplify the complicated loading spectra encountered in practice, two extreme electro-mechanical loading states are considered in this work: in-phase and out-of-phase. An example of in-phase loading is shown in Fig. 1(Left): A loading of the E field from -0.4 to $+2$ kV/mm is accompanied by a synchronous loading of the compressive stress up to a specific maximum magnitude. Following that, the electric field and stress are unloaded simultaneously. That is, the two external loads go through their maximum and minimum points at the same time and in the same direction (using absolute value for stress loading). As shown in Fig. 1(Right), the out-of-phase loading means a loading of the stress is accompanied by an unloading of the E field from its positive peak or do it the other way round. That is, the stress loading acts 90° later in time than does the electric field. Notice that the phase relationship defined in this work is consistent with physical convention, whereas a contrary definition was used in the work of Mitrovic et al.⁹

3. Results and discussion

3.1. Polarization and strain response under loading with zero-maximum stress

An example of polarization and strain versus electric field response under released compression (zero-maximum stress) loading condition is presented in Fig. 2 for a stress range of 60 MPa. The results obtained in the stress-free state are also given for comparison.

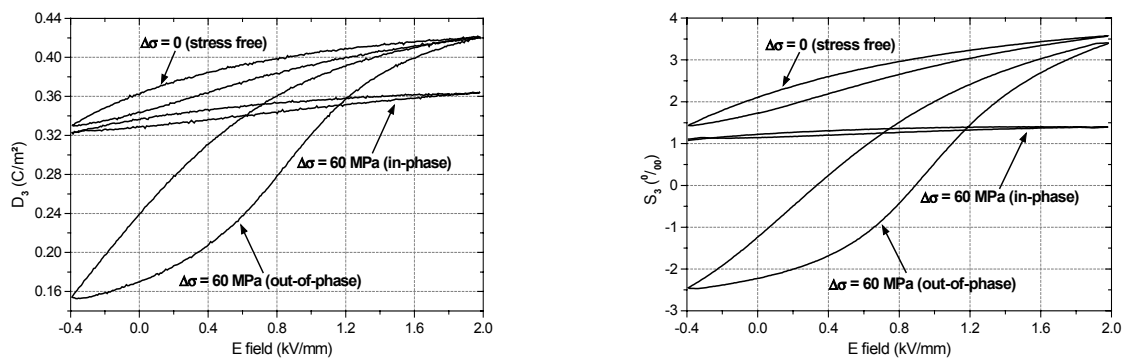


Fig. 2. Comparison of the polarization (Left) and strain (Right) behavior in the stress free state, under in-phase and out-of-phase electromechanical loading with zero-maximum stress.

Even in the stress-free state, the P-E and S-E curves do not show ideally linear responses, but exhibit pronounced hysteresis and non-linearity. This is due to some amount of domain alignment repeatedly induced by the cyclic E field loading. Zhang et al.⁶ proved that at room temperature and under stress-free conditions, extrinsic non-180° domain switching made the major contribution to the dielectric and piezoelectric responses of PZT ceramic materials.

When the mechanical load is applied in-phase with electrical load, increasing of the E field from -0.4 to +2 kV/mm is accompanied by a loading of the stress from zero to the maximum magnitude of -60 MPa. As shown in Eq. 1-2, the compressive stress induced direct piezoeffect ($d_{33}\sigma_3$) and elastic deformation ($s_{33}\sigma_3$) will cause a decrease in the polarization and strain, respectively. In addition, the synchronously increasing compression load will act against the E field to prevent domain switching to the poling direction, by which a gradual reduction of P_3^r , S_3^r , d_{33} , and ϵ_{33} will be induced. As a result of the combined action, we find the polarization and strain responses are significantly impeded, and the curves display slight hysteresis.

A completely contrary phenomenon is observed for the specimen exposed to out-of-phase electro-mechanical loading. In such a case, unloading of the E field from +2 to -0.4 kV/mm is accompanied by increasing stress load from zero up to the maximum magnitude. Their combination will induce large amount of non-180° domain switching perpendicular to the poling direction. As a result, both polarization and strain are observed to decrease drastically (including the aforementioned effects of direct piezoeffect and elastic deformation). Following that, loading of the E field together with unloading of the stress will gradually reorient most of the domains back to the poling direction. When the E field reaches +2 kV/mm, the stress returns to zero, the polarization and strain are almost recovered to the same values in the stress-free state. Due to large amount of domain switching induced during the loading-unloading process, large hysteresis and non-linearity are observed in the polarization and strain curves. We can anticipate that more pronounced hysteresis will be observed with the stress range increment.

The measurements were repeated for different stress ranges of up to 150 MPa. The polarization and strain differences between +2 and -0.4 kV/mm were plotted in Fig. 3 as a function of stress range. In the case of in-phase loading, the polarization and strain outputs monotonically decrease with an increase in load range. At 75 MPa, the mechanical loading completely eliminates the piezoelectric strain (strain induced by E field). After it, the strain output becomes negative, which indicates the strain behavior is mainly determined by mechanical elastic deformation. An inverse effect occurs when the specimen subjected to out-of-phase electro-mechanical loading, the polarization and strain outputs increase with increasing stress range and tend to be saturated at high range levels.

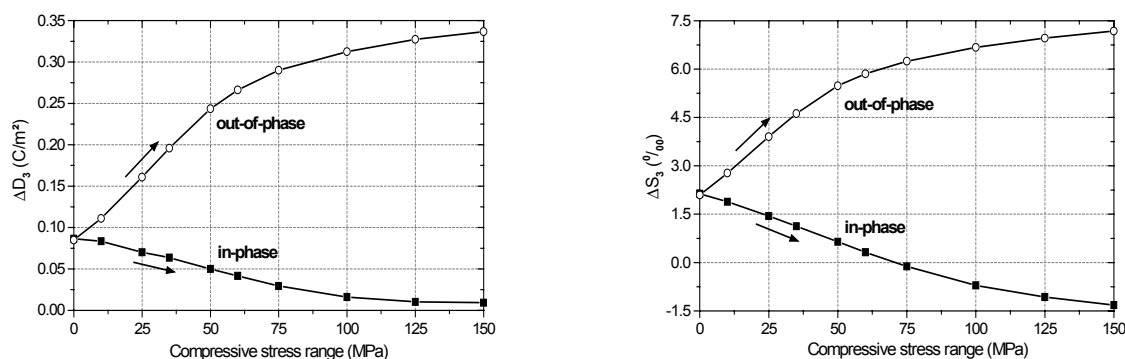


Fig. 3. Polarization (Left) and strain (Right) output as a function of stress range for in-phase and out-of-phase electro-mechanical loading with zero-maximum stress.

3.2. Polarization and strain response under loading with constant mean stress

Figs. 4-7 show the polarization and strain response under cyclic mechanical load with a constant-mean stress of -25 MPa. The range of stress amplitude is between 0 and 25 MPa, with an increment of 5 MPa between two steps. The curves are plotted versus electric field and mechanical load, respectively.

In Figs. 4-5, mechanical load of different amplitudes is applied in-phase with electrical load. In such a case, loading of the E field from -0.4 to +2 kV/mm is accompanied by a synchronous increase in compressive stress, which will act against the electric field to inhibit

domain alignment to the poling direction. With stress amplitude σ_a increment, more and more domains will be constrained in the perpendicular direction and can't be oriented by the electric field to provide contributions to the polarization and strain, the hysteresis of the P-E and S-E curves therefore becomes less and less pronounced.

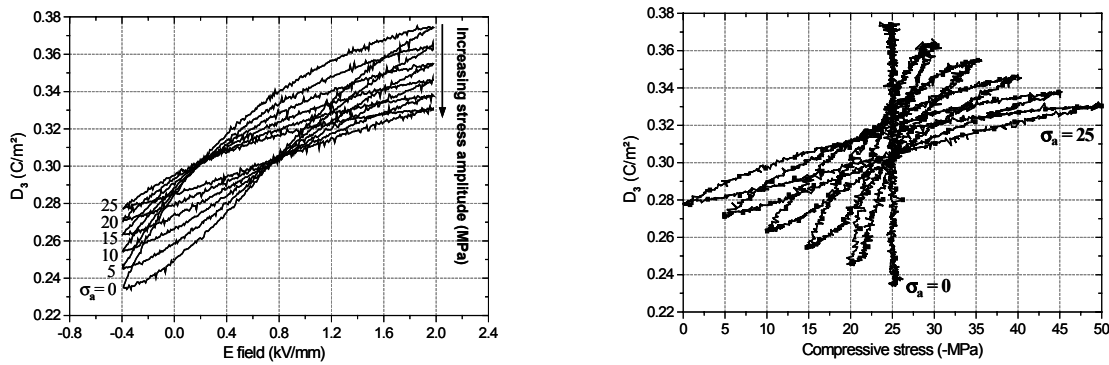


Fig. 4. In-phase electro-mechanical loading with -25 MPa constant-mean stress: Polarization response as a function of electric field (Left) and mechanical load (Right).

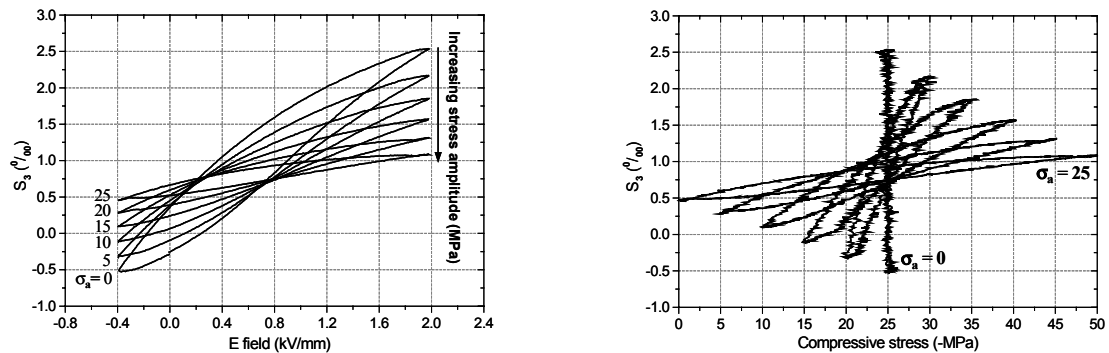


Fig. 5. In-phase electro-mechanical loading with -25 MPa constant-mean stress: Strain response as a function of electric field (Left) and mechanical load (Right).

Figs.6-7 correspond to the out-of-phase electric-mechanical loading. As mentioned before, during the unloading period of E field from $+2$ to -0.4 kV/mm, a gradually increasing compressive stress will induce non- 180° ferroelastic domain switching. A subsequent reloading of the E field with decreasing stress will reorient more domains switching back to the poling direction. As a result, an enhancement of the polarization and stress response with larger hysteresis is observed with σ_a increment.

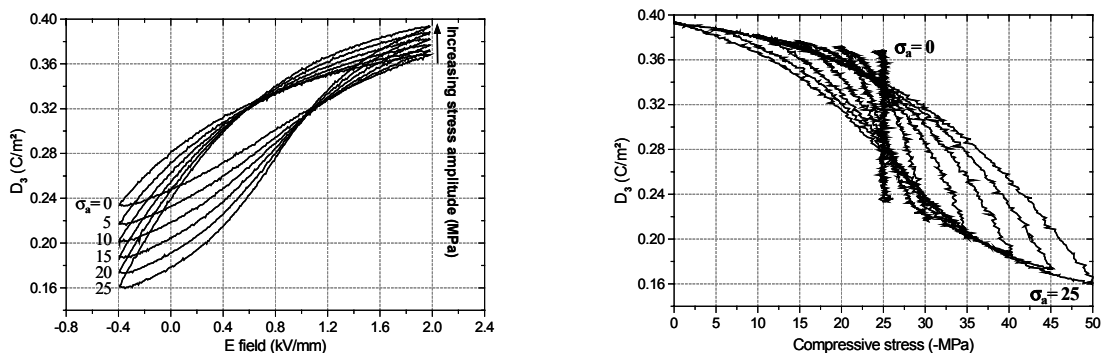


Fig. 6. Out-of-phase electro-mechanical loading with -25 MPa constant-mean stress: Polarization response as a function of electric field (Left) and mechanical load (Right).

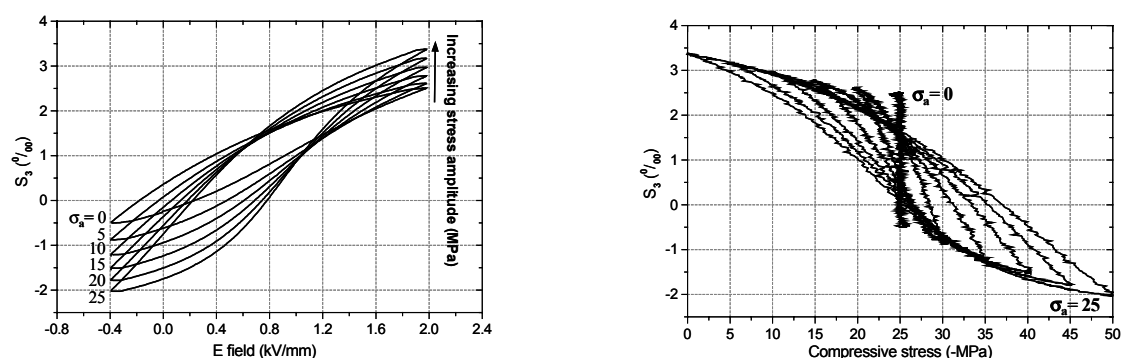


Fig. 7. Out-of-phase electro-mechanical loading with -25 MPa constant-mean stress: Strain response as a function of electric field (Left) and mechanical load (Right).

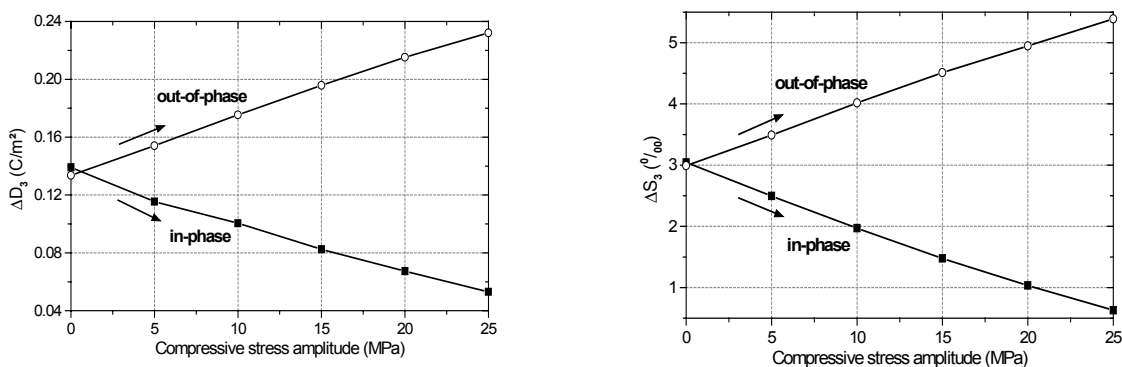


Fig. 8. Polarization (Left) and strain (Right) output as a function of stress amplitude for in-phase and out-of-phase electro-mechanical loading with -25 MPa constant-mean stress.

The polarization and strain outputs between $+2$ and -0.4 kV/mm are measured and plotted in Fig. 8 as a function of stress amplitude. In the case of in-phase loading, ΔD_3 and ΔS_3 linearly decrease with increasing stress amplitude. An inverse effect occurs for the out-of-phase

loading, as expected, both ΔD_3 and ΔS_3 monotonically increase with an increase in stress amplitude σ_a .

The measurements were repeated for a constant-mean stress of -50 MPa. The load amplitude ranges between 0 and 50 MPa, with an increment of 5 MPa between two steps. Only the strain response is shown in Fig. 9 for in-phase electro-mechanical loading, because the actuation behavior is more interesting for a designer and the in-phase loading represents a worst case of operating scenario.

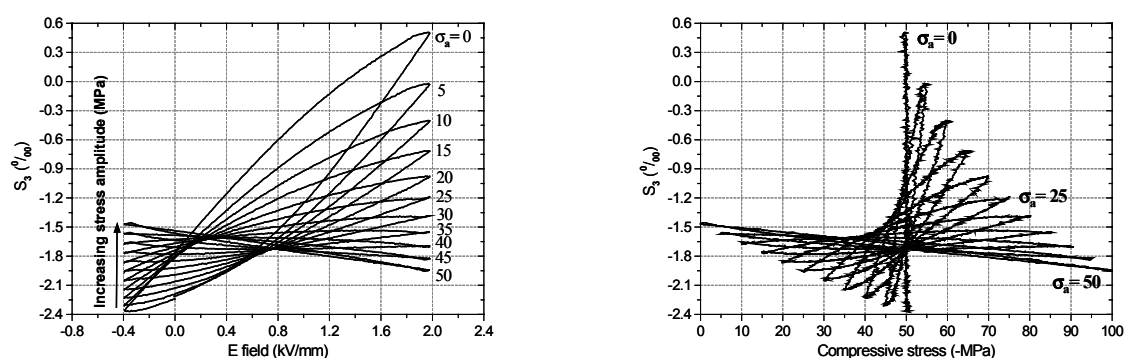


Fig. 9. In-phase electro-mechanical loading with -50 MPa constant-mean stress: Strain response as a function of electric field (Left) and mechanical load (Right).

Similar to the behavior at -25 MPa mean stress, Fig. 9 clearly demonstrates a suppression of the strain response for a specimen undergoing in-phase electro-mechanical loading. The actuation output and hysteresis of S-E curves decrease quickly with an increase in load amplitude. At about $\sigma_a = 40$ MPa, the piezoelectric strain is completely eliminated by the mechanical load and, the strain output becomes negative at higher stress amplitude levels.

So far, the polarization and strain responses of piezoceramics under several alternating stress loading conditions are presented. Keep in mind that the real electric field/stress-time pattern encountered in practical actuator applications can be more complicated. The active material may experience asynchronous electro-mechanical loading, various loading frequencies, random or periodical changing of mean stress and amplitude, etc. The loading spectra

investigated in this work are beginning to approach a degree of realism, the experimental results may therefore serve as a basis for the design of piezoelectric actuators.

4. Summary and conclusions

In this experimental work, high-field induced polarization and strain responses were measured for a commercial soft PZT material under cyclic compressive stress loading with different mean stresses and amplitudes. When the stress is applied in-phase with electrical load, a suppression of the dielectric and actuation response is observed due to the mechanical load impedes domain switching to the poling direction. However, applying an out-of-phase mechanical load has the adverse effect on the polarization and strain response since more domain switching induced during the loading-unloading period of electric field, the overall strain output is therefore significantly improved with increasing hysteresis and nonlinearity.

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